

Understanding and Improving Pinning in Coated Conductors

Part II: Improving Pinning

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Plans which were put forward for 2004

Design and implement systematic experiments to determine if chemical modifications to REBCO offer enhanced performance, particularly in an external magnetic field.

Goal: Reproducibly double J_c at 75 K in a magnetic field parallel to the c-axis.

To grow HTS films with rare earth substitutions.

Goal: to search for pinning enhancements by introduction of random defects.

To introduce columnar defects at different angles in YBCO films on single crystal substrates.

Goal: to measure angular dependence of J_c in samples with a controlled defect structure, for comparison with CC.

Possible practical ways to enhance pinning

Examples of earlier work relevant to this study

- More line defects
 - Miscut substrates *Lowndes D.H. et al. Phys. Rev. Lett. 74 (12) 2355 (1995)*
 - Lower growth temperatures (smaller island sizes) *Dam B. et al. Phys. Rev. B 65 (6) 064528 (2002)*
 - Heteroepitaxial nanoparticles *Huijbregtse J. M., et al., Phys. Rev. B. 62 (2) 1338-1349 (2000)*
- Point defects (and associated strain)
 - cation or anion vacancies
 - RE-Ba cross substitution
- Volume defects (and associated strain)
 - Second phase particles *Berenov A. et al. J. Mater. Res. 18 956 (2003)*
 - Interlayers of non-superconducting material *Haugan T. et al., J. Mater. Res. 18 (11) 2618 (2003)*
 - Substrate surface roughening *Jia Q.X. et al., APL 80 (9) 1601 (2002)*

Four different routes to enhanced pinning demonstrated

Method 1. Change RE ion size variance

- introduces random point defects

Method 2. Change average RE ion size

- introduces random points defects and correlated defects

Method 3. Introduction of buffer surface roughness

- more low angle grain boundaries || c

Method 4. Introduction of heteroepitaxial second phases

- increase c-axis dislocation density

Substrates used were single crystal SrTiO_3 , SrTiO_3 -buffered single crystal MgO and SrTiO_3 -buffered IBAD MgO. Films around 1-1.5 μm thick. Growth by PLD, standard YBCO conditions used for all samples.

Methods 1 and 2: Applying a systematic approach to studying mixed REBCO's.

- The aim is to apply a systematic approach to mixing RE ions.
(Changing growth T , pO_2 , or mixing ratio are not structurally systematic)
- We deliberately paid no attention to increasing T_c above that for YBCO because much higher (and therefore not practical) growth temperatures are required for the higher T_c RE's. All our samples had the same or lower T_c than YBCO.
- We studied the influence of RE ion size variance and RE ion size separately

Method 1: Change RE ion size variance for constant RE ion size

Method 2: Change RE ion size for constant RE ion size variance

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Method 1. Change RE ion size variance

- introduces random point defects

Method 2. Change average RE ion size

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Method 3. Introduction of buffer surface roughness

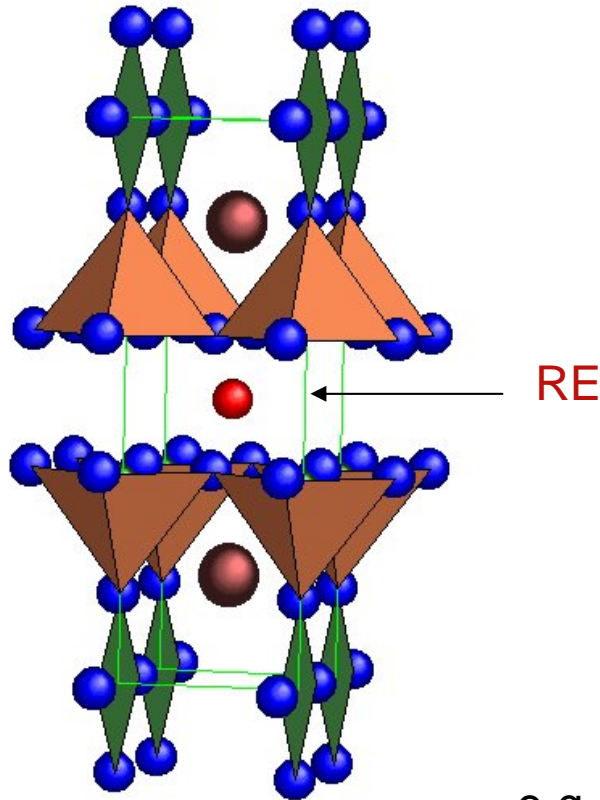
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Change ion size variance, and keep mean ionic radius constant



$$\sigma^2 = \sum y_i \langle r_i \rangle^2 - \langle r_A \rangle^2$$

where σ^2 is the variance of the mixture of RE ions i

y_i is the mole fraction of ion i

r_i is the ionic radius of ion i

r_A is the mean ionic radius

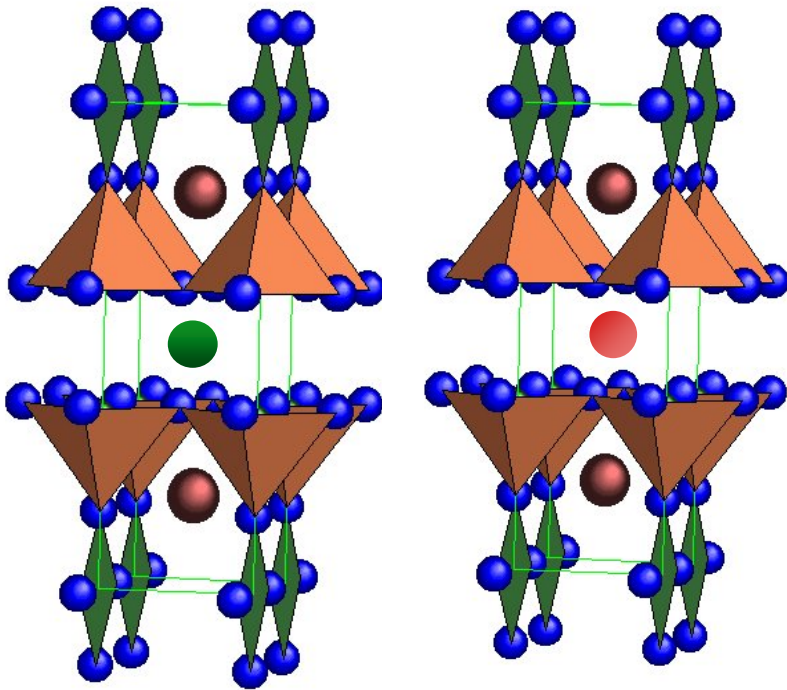
$\langle r_A \rangle = 1.019 \text{ \AA}$, i.e. the ionic radius was kept constant at the size of Y^{3+}

e.g. for the composition $\text{Dy}_{1/3}\text{Ho}_{2/3}\text{BCO}$

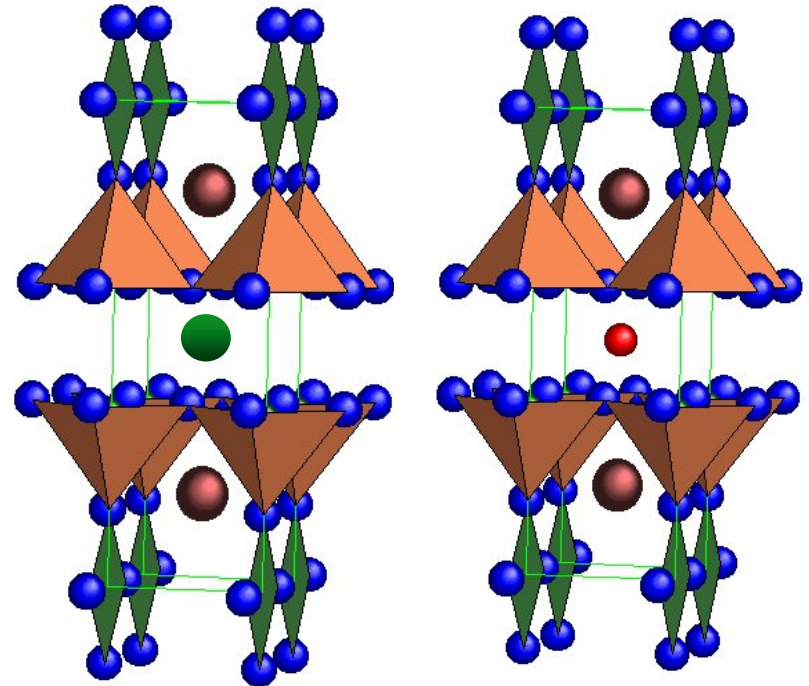
$$\sigma^2 = [1/3(1.027^2) + 2/3(1.015^2)] - 1.019^2 = 0.32 \times 10^{-4} \text{ \AA}^2$$

Method 1: Change RE ion size variance

Aim is to produce random variations in oxygen ion displacements



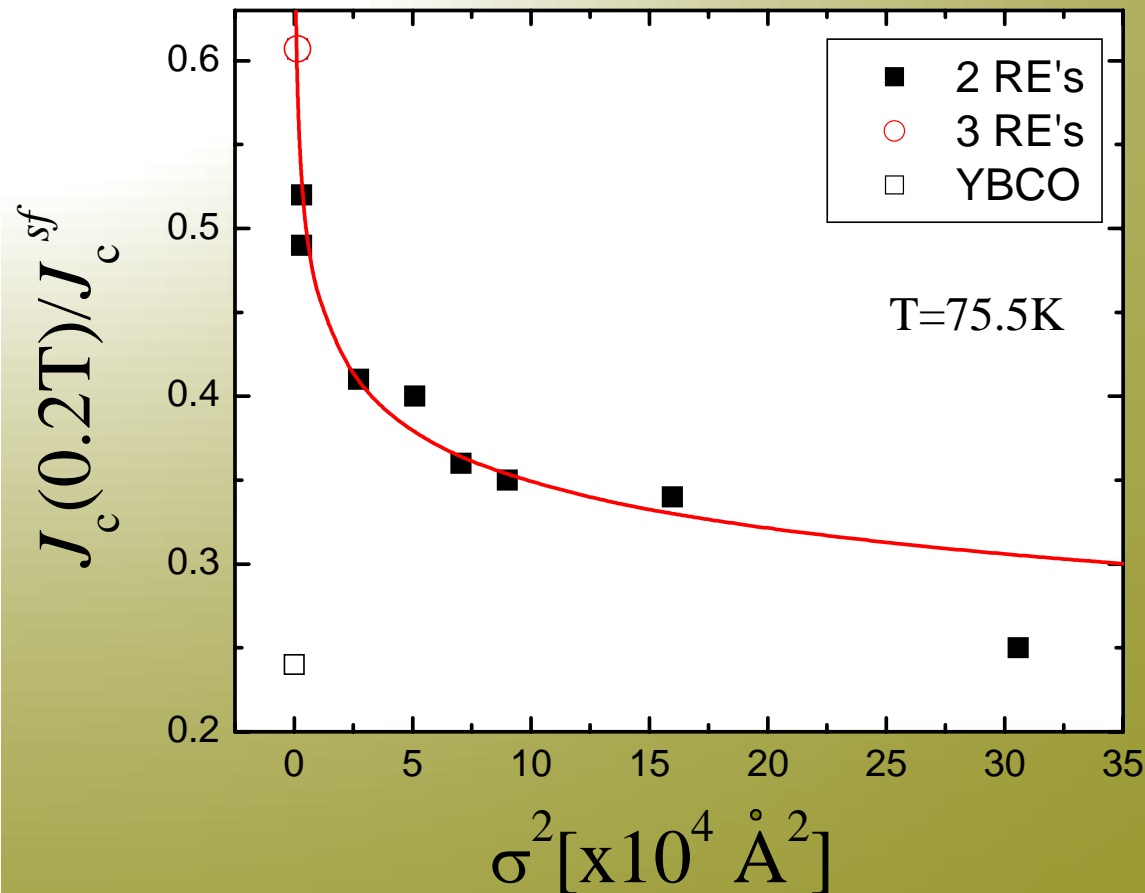
small variance



large variance

Changing RE ion size does not change Cu(2)-O(2) plane buckling
but increases the Cu(2)-O(1) distance by ~2.5% across the RE series

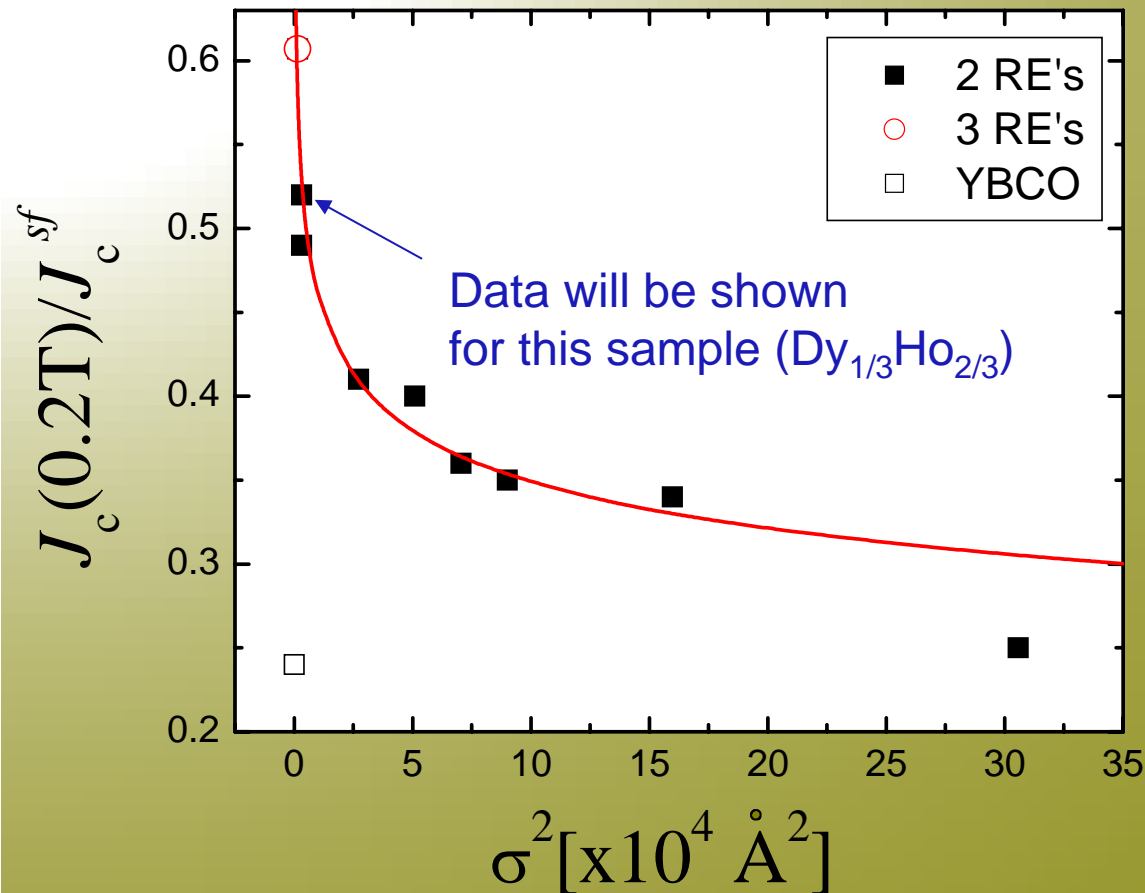
There is a Systematic, Reproducible, and Improved Dependence of Low Field Pinning on RE Ion Size Variance



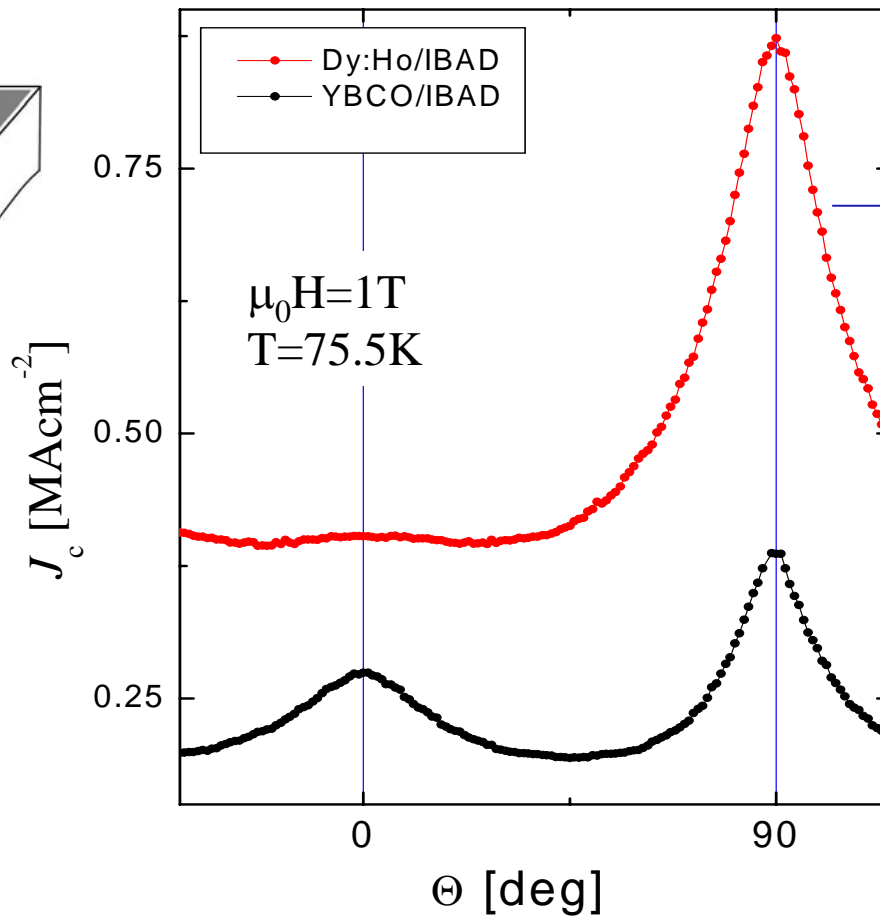
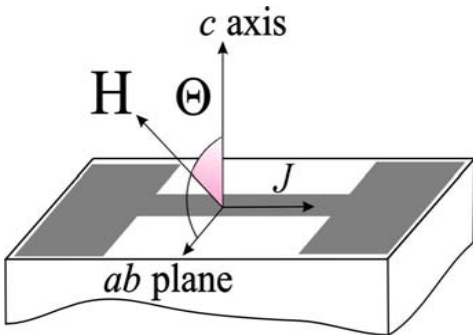
The variance effect is *intrinsic* and hence should work for any processing route.

J.L. MacManus-Driscoll et al. APL **84** (26) (2004) 5329

There is a Systematic and Reproducible Dependence of Low Field Pinning on RE Ion Size Variance

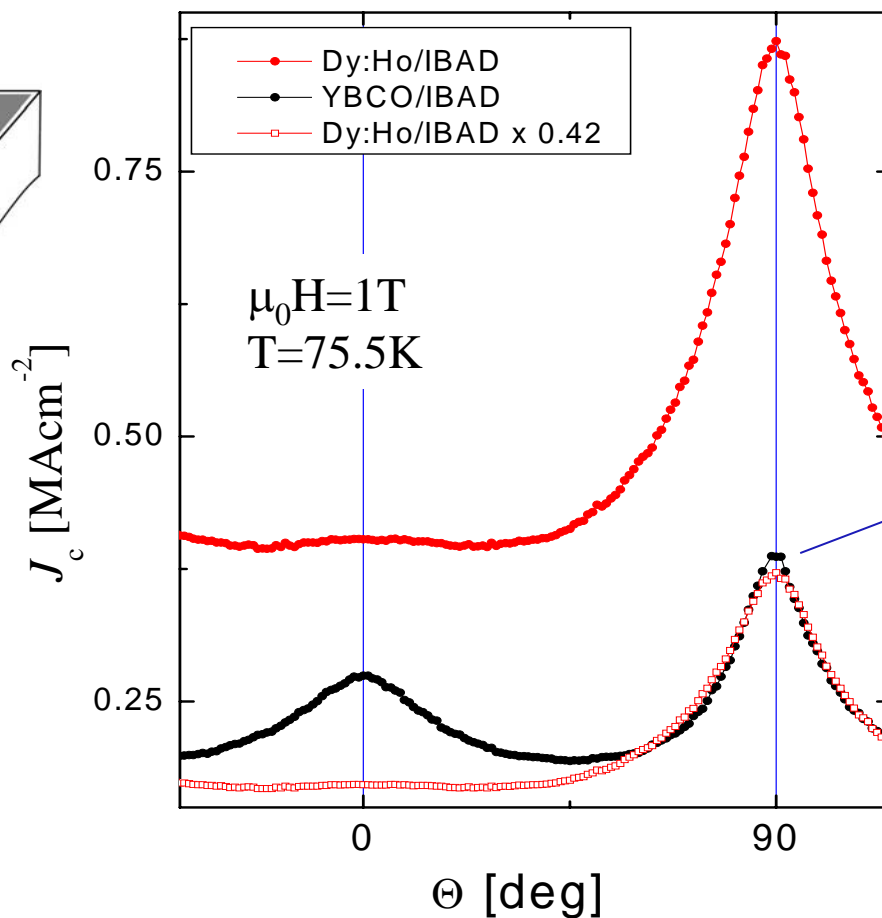
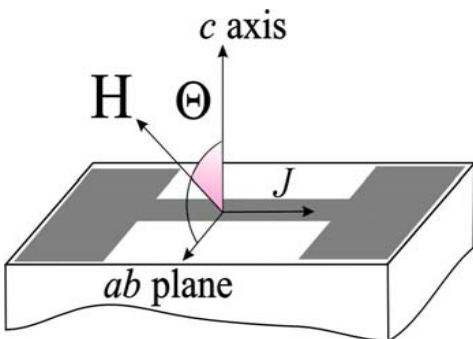


Field angular measurements indicate that random, point-like defects responsible for increased J_c



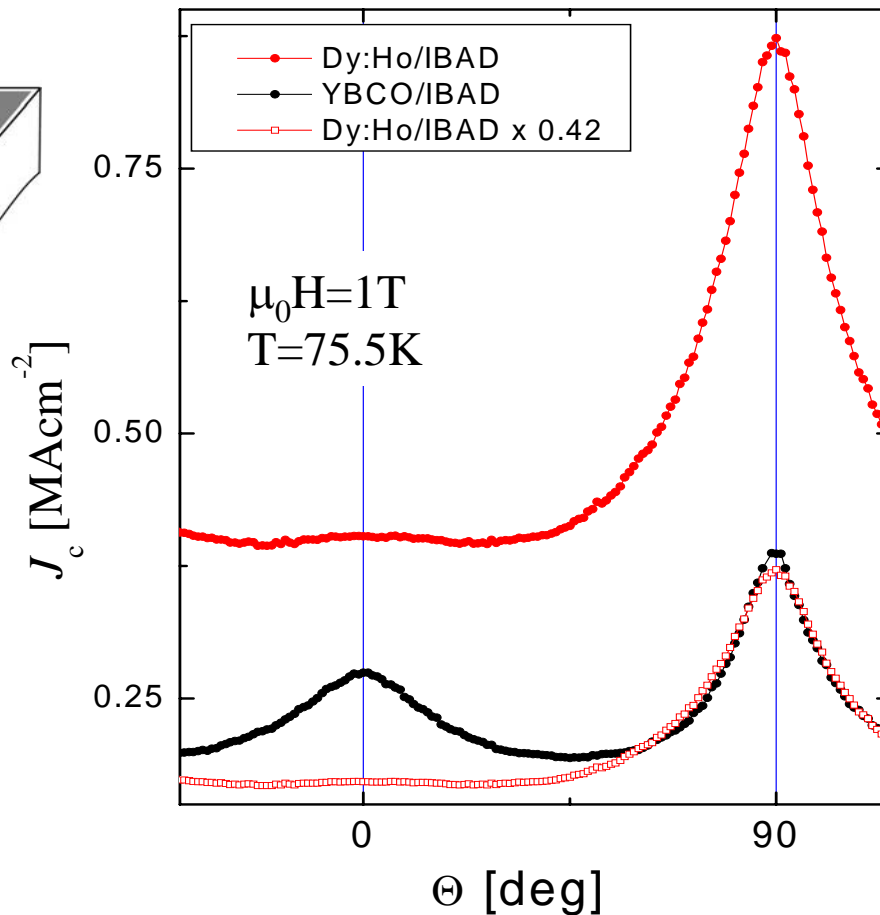
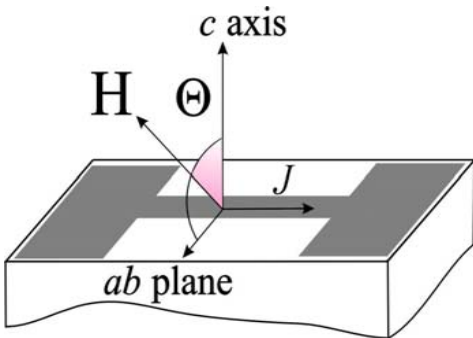
Optimum variance sample has improved J_c over wide field and angular range

Field angular measurements indicate that random, point-like defects responsible for increased J_c



Normalisation indicates non-correlated, random, point-like defects present

Field angular measurements indicate that random, point-like defects responsible for increased J_c

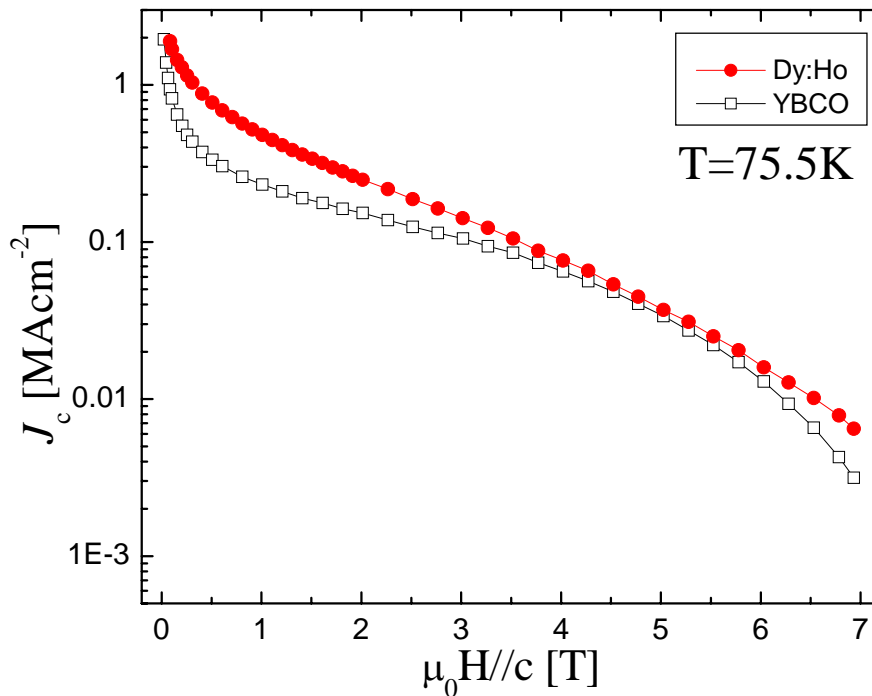


The results are consistent with earlier studies on magnetoresistive and ferroelectric perovskites \Rightarrow Ion size variance produces *random* displacements of oxygen ions

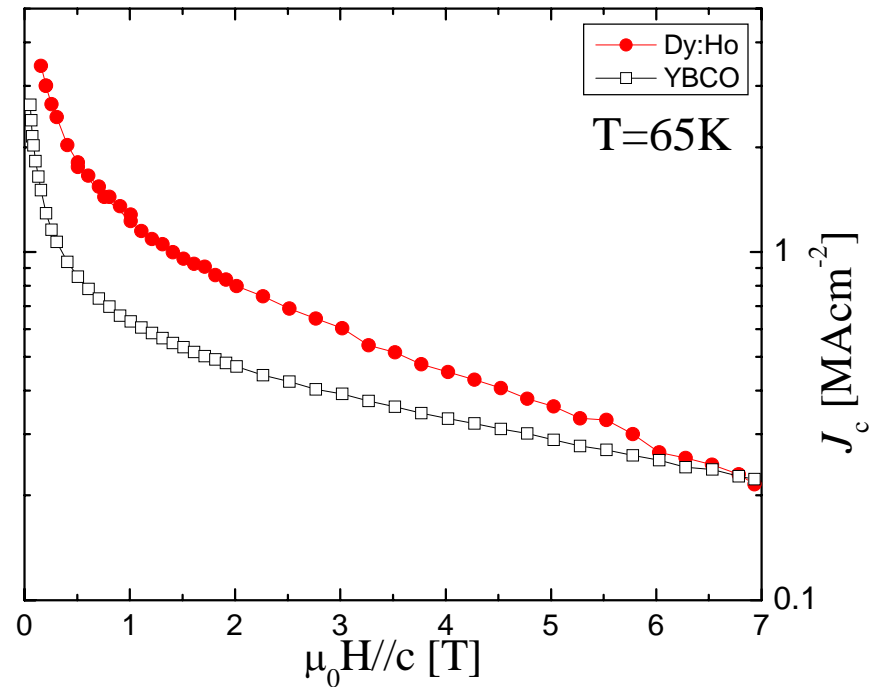
(L. M. Rodriguez- Martinez, and J.P. Attfield, Phys. Rev. B 54 (22) (1996) R15 622)

J_c versus field improved by up to a factor of 2 for lowest variance sample

Low ion size variance improves
field dependence to 4T at 75.5K



Low ion size variance improves
field dependence to 6T at 65K



Four different routes to enhanced pinning demonstrated

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Method 2. Change average RE ion size

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Method 3. Introduction of buffer surface roughness

- more low angle grain boundaries || c

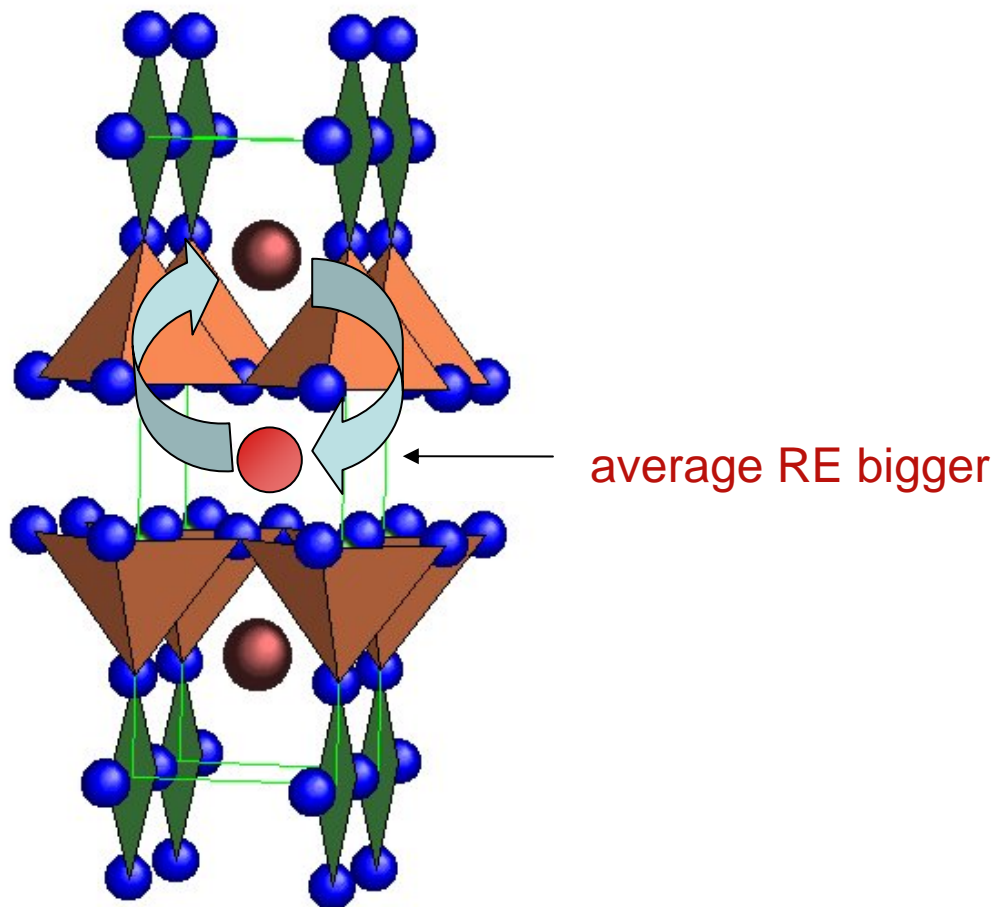
Method 4. Introduction of heteroepitaxial second phases

- increase c-axis dislocation density

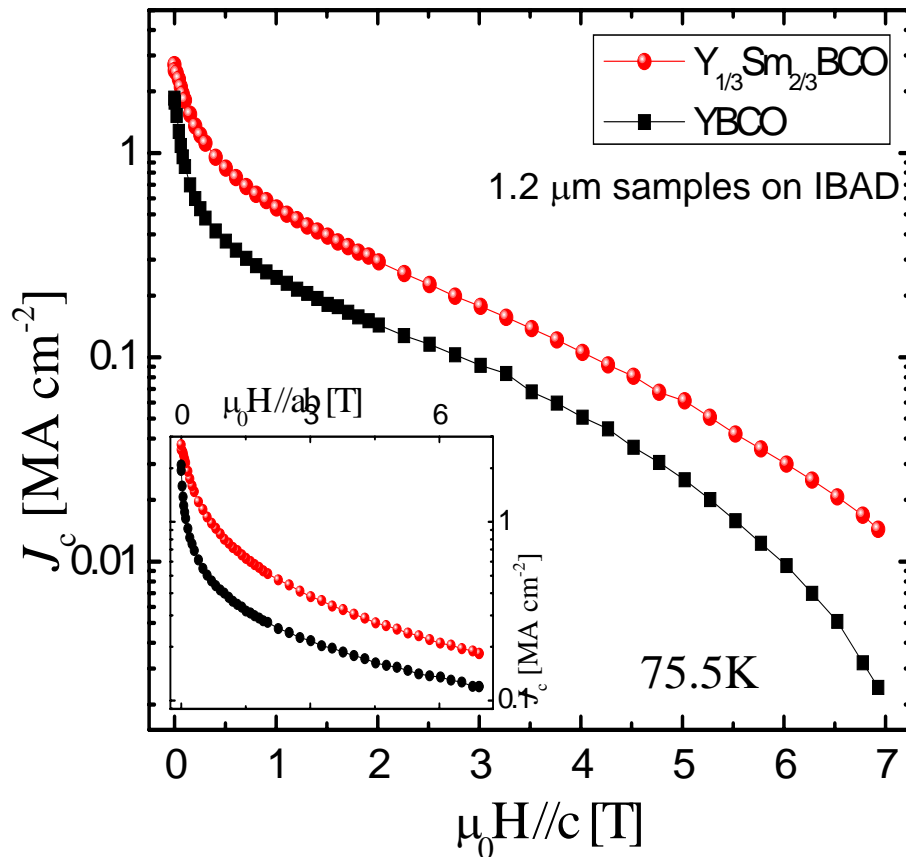
Substrates used were single crystal SrTiO_3 , SrTiO_3 -buffered single crystal MgO and SrTiO_3 -buffered IBAD MgO. Films around 1-1.5 μm thick. Growth by PLD, standard YBCO conditions used for all samples.

Method 2: Increase average RE ion size, keep variance constant

We find no systematic dependence of pinning on RE ion size. This is not surprising since the amount of Y-Ba cross substitution is strongly dependent on kinetics which are not easily controlled.



J_c of $Y_{1/3}Sm_{2/3}BCO$ ($\langle r_A \rangle = 1.039 \text{ \AA}$) is much improved compared to YBCO ($\langle r_A \rangle = 1.019 \text{ \AA}$). $\sigma^2 = 6-8 \times 10^{-4} \text{ \AA}^2$



On STO: Self-field J_c up to 5 MA cm^{-2} in $1 \mu\text{m}$ thick films.

On IBAD: $J_c(H)$ improved by a factor of 2-10 (depending on H)

The RE ion size effect is *intrinsic* and hence should work for any processing route.

J.L. MacManus-Driscoll et al., submitted APL 6/04

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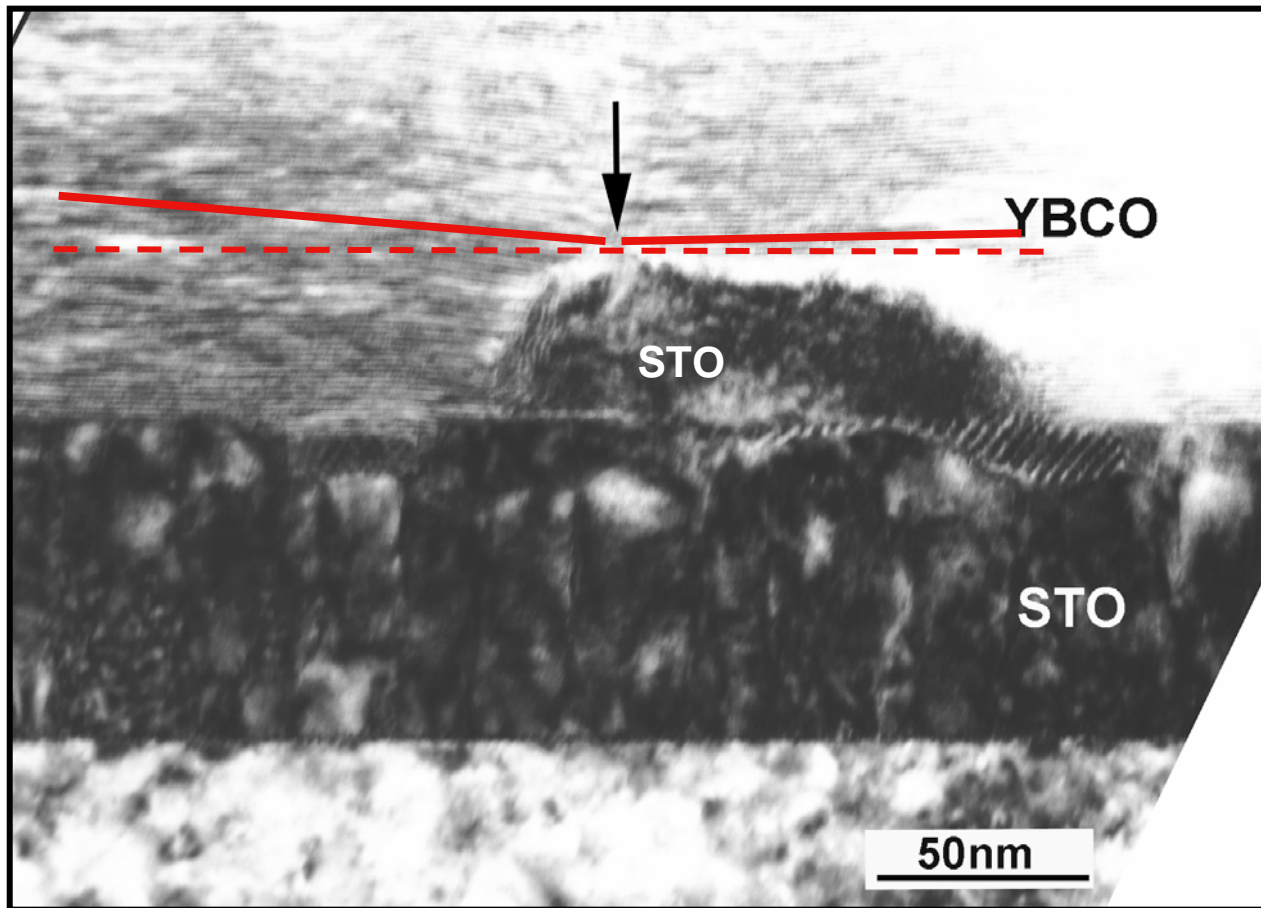
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Method 3: Introduction of buffer surface roughness by lower temperature growth introduces low angle grain boundaries



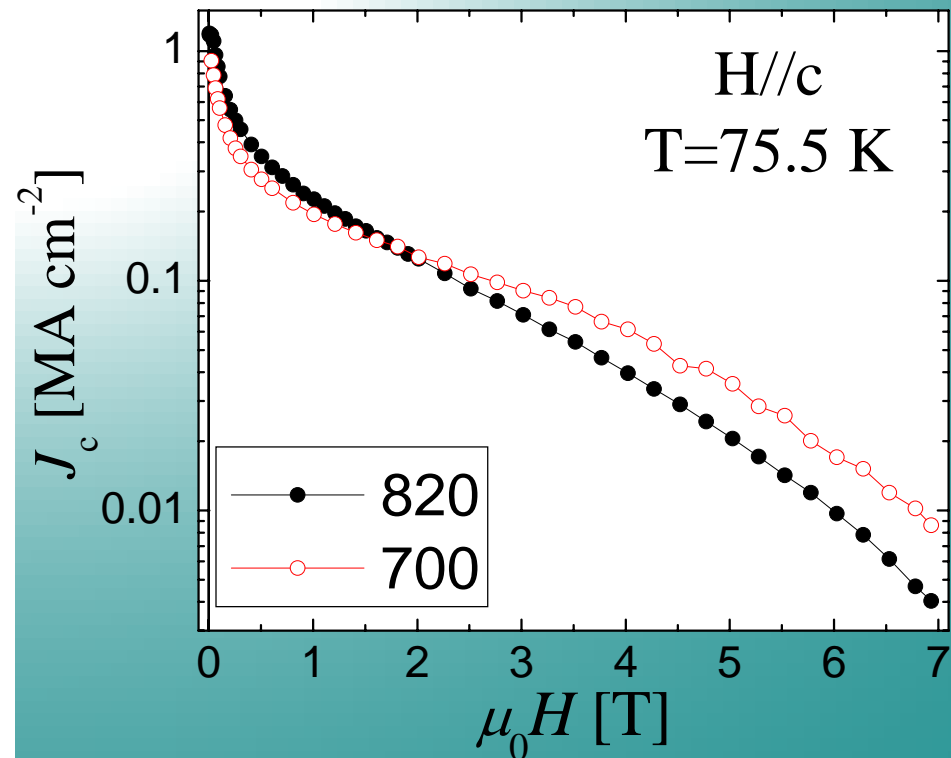
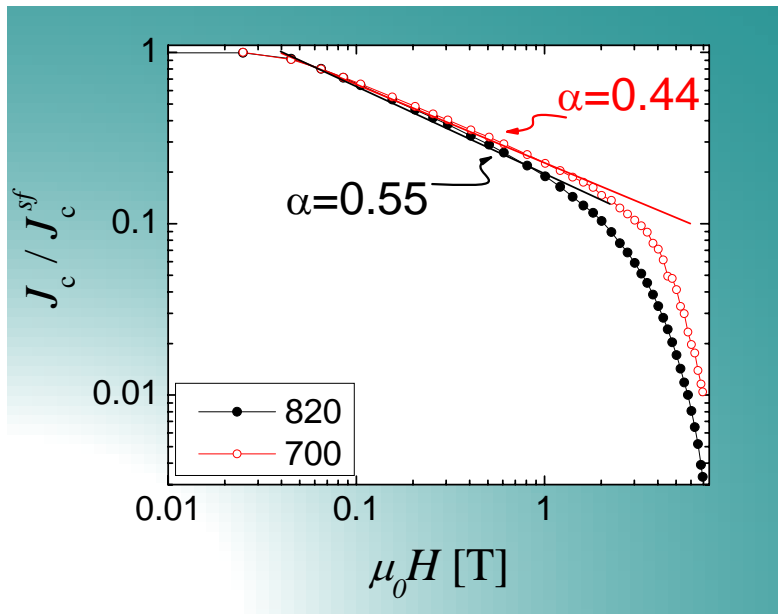
H. Wang et al. J. Mat. Res. 19 (2004) 1869

Superconductivity for Electric Systems - Annual Peer Review ♦ July 27-29, 2004 ♦ Washington DC

The *in-field* dependence of J_c is improved in 5 μm thick films on IBAD-MgO

$T_{\text{STO}} = 700\text{ }^\circ\text{C}$ cf. $T_{\text{STO}} = 820\text{ }^\circ\text{C}$

- Lower J_c self-field
- Higher J_c in-field

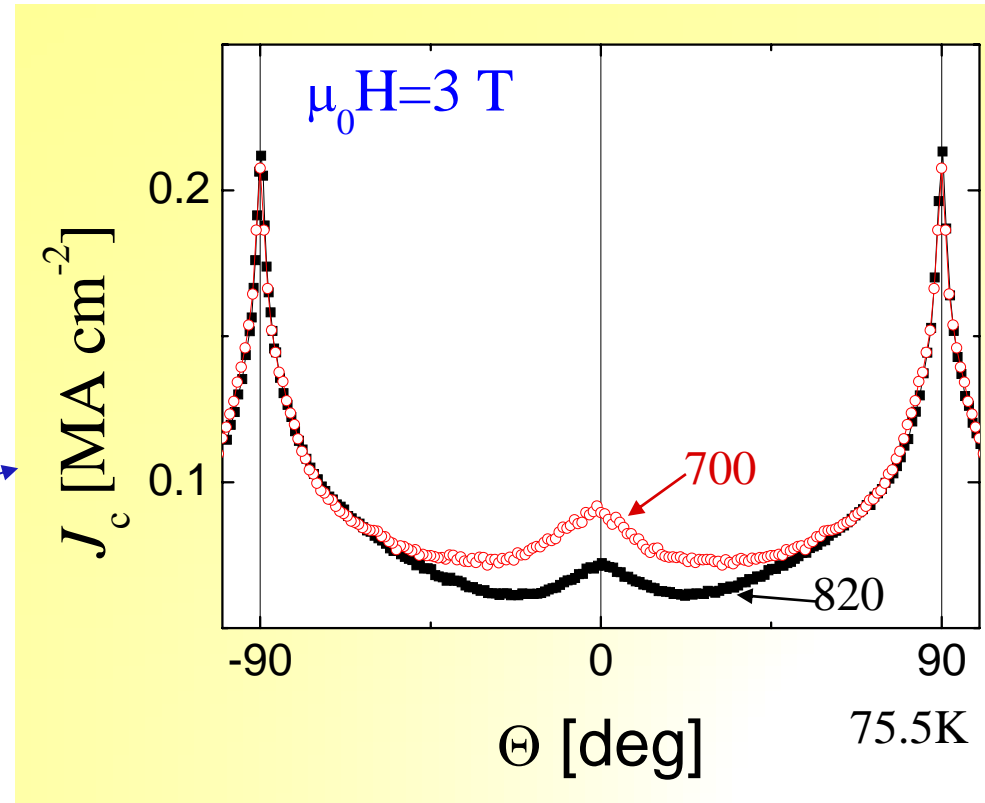
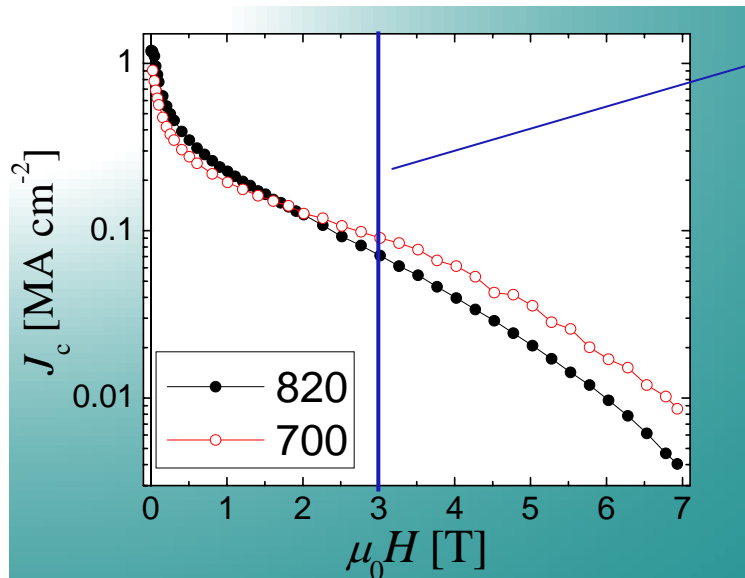


Growth temperature of YBCO was constant at 760°C

B. Maierov et al. accepted Ceramic Transactions (2004)

The angular dependence of J_c shows a larger c-axis peak

The angular effect is consistent with the presence of low angle boundaries //c (introduced by tilted a-b planes induced by buffer surface roughness)



*B. Maiorov et al.
accepted Ceramic Transactions (2004)*

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Introduction of c-axis dislocations through heteroepitaxial second phases

Materials selection for second phase:

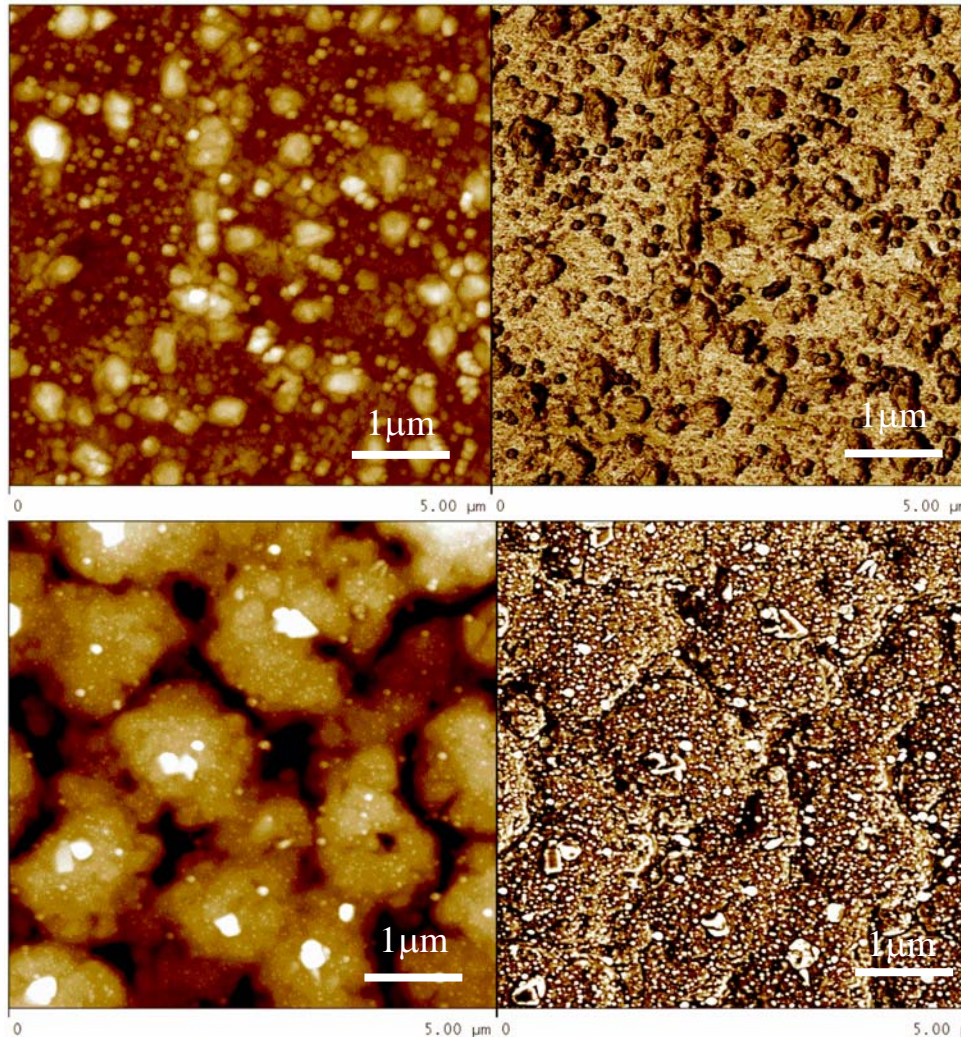
- a) Can grow heteroepitaxially with YBCO
- b) Lattice mismatch producing strain leading to misfit dislocations
- c) High melting temperature phase, yielding slow growth kinetics and hence small particles
- d) Chemical compatability with YBCO

Simple method of second-phase incorporation:

- ceramic target of YBCO+ 5 mol% BaZrO₃ fabricated and ablated

J.L. MacManus-Driscoll et al. Nature Materials 3 (7 July) (2004) 439

Micrographs of 'YBCO+BaZrO₃' shows the presence of surface nano-particles

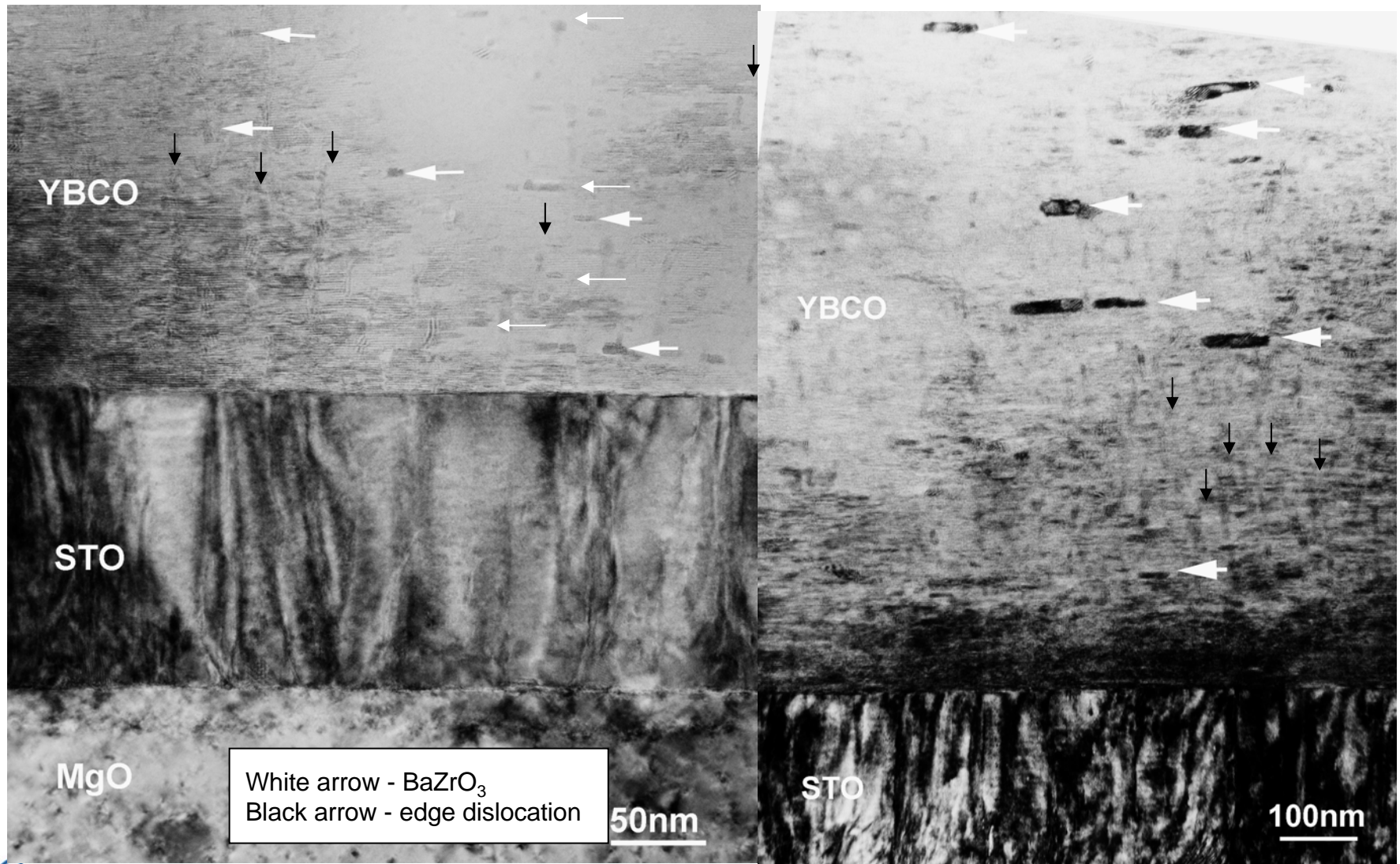


YBCO

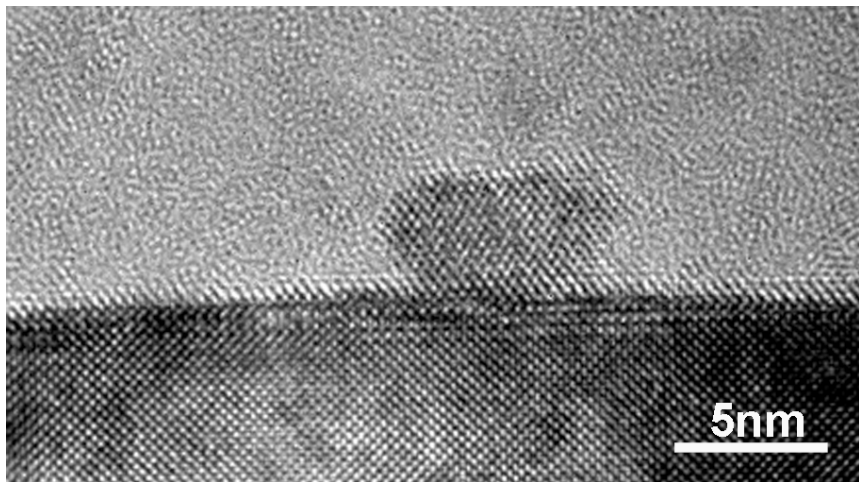
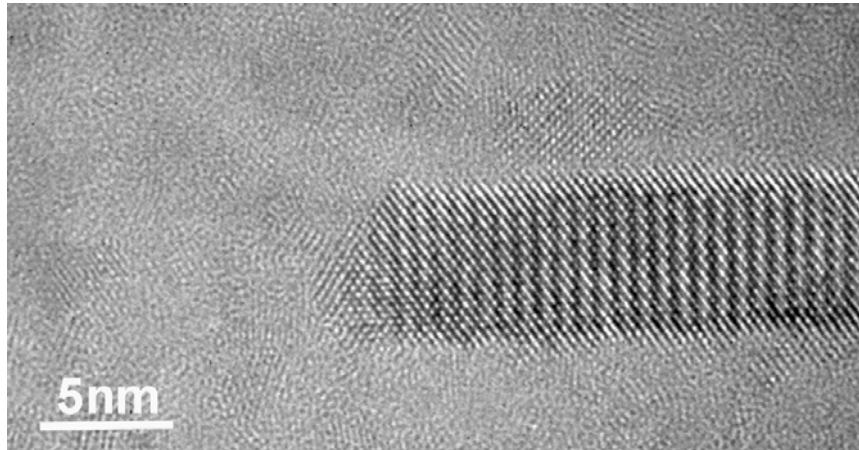
YBCO+BaZrO₃

10-100nm surface particles. In bulk too?

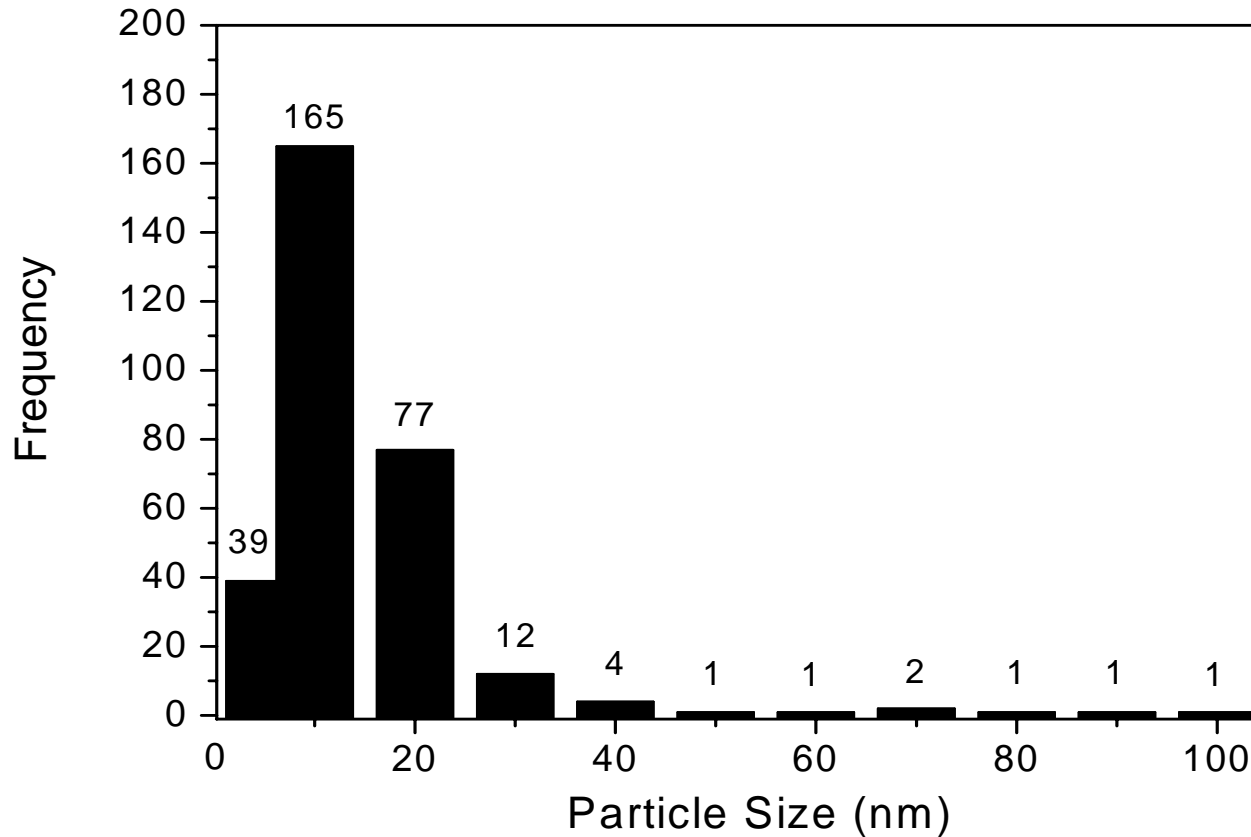
TEM shows edge dislocations. Around a 5-fold increase in density of c-axis dislocations ($\sim 400 \mu\text{m}^{-2}$ cf. $\sim 80 \mu\text{m}^{-2}$)



HRTEM and EDX shows the presence of BaZrO_3 nano-particles embedded within the film

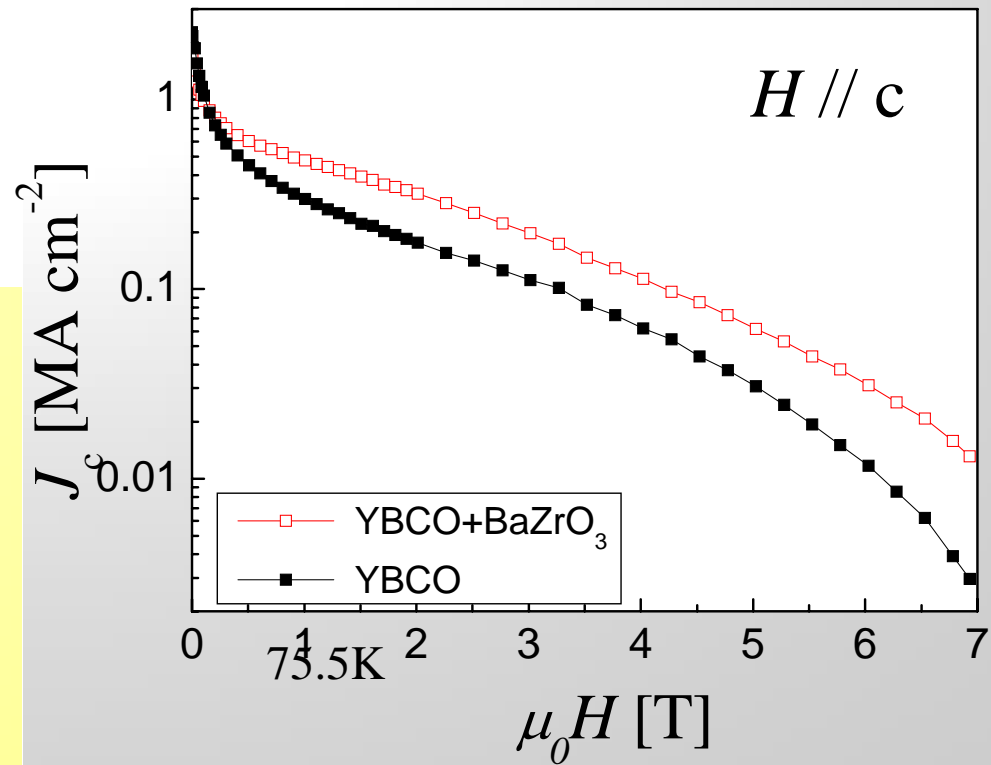
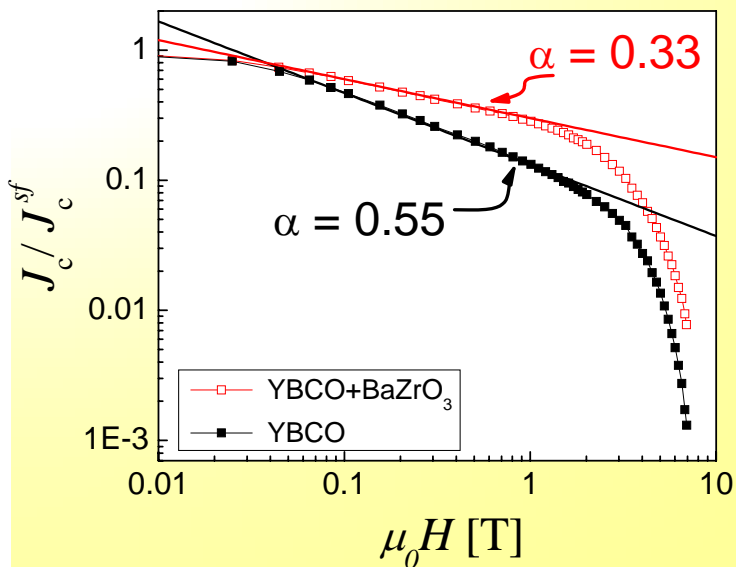


Modal particle size of BaZrO_3 is 10 nm

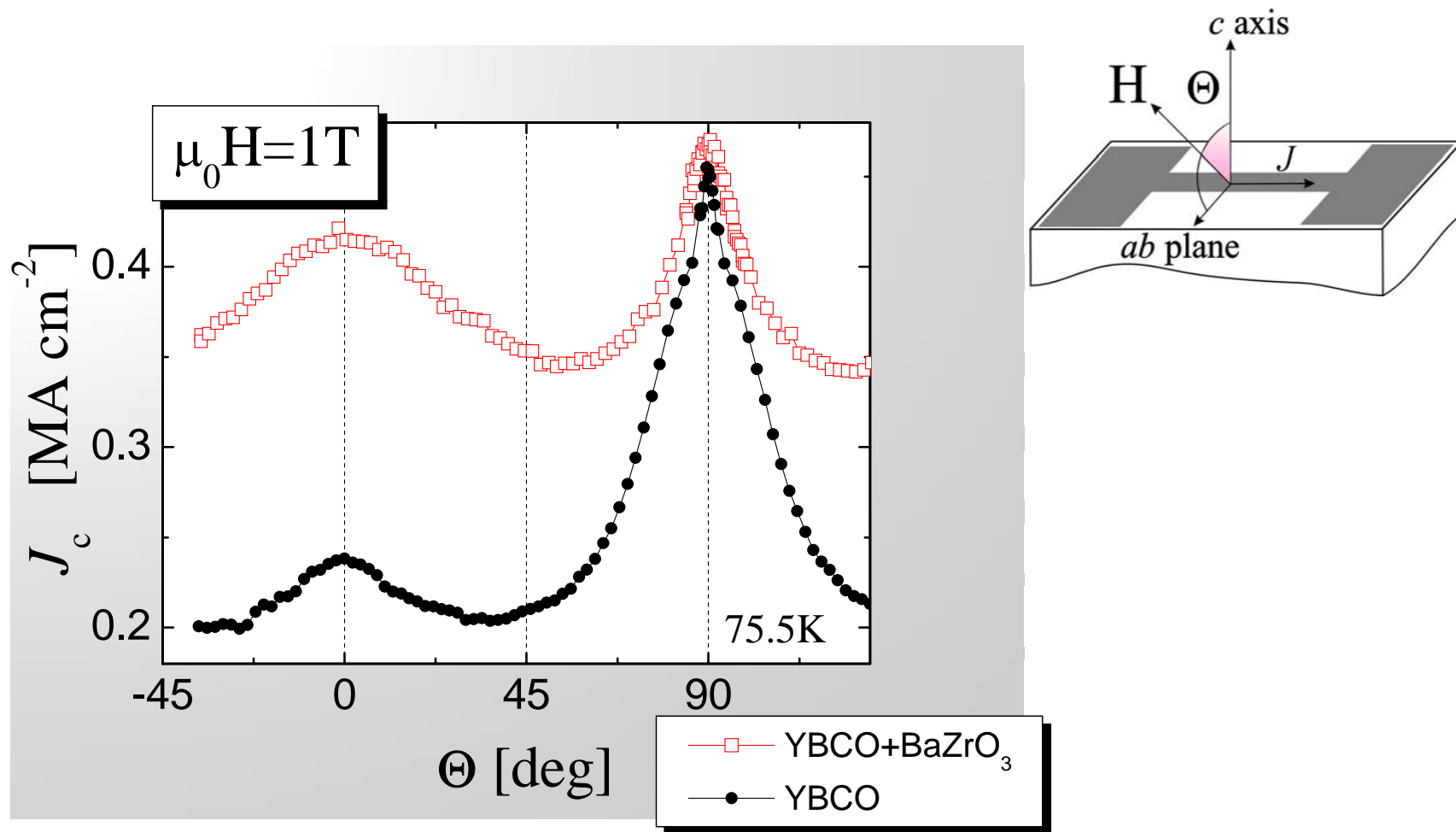


J_c vs. $\mu_0 H$ comparison of YBCO+BaZrO₃ and YBCO samples shows a 1.5-5x increase in J_c

The ' α ' value is much lower for YBCO+BaZrO₃



Field angular data shows a huge c-axis peak consistent with the additional c-axis dislocations



Conclusions

- Several successful routes have been demonstrated to nano-engineer defects into REBCO to enhance pinning.
 - simple, inexpensive and scaleable technique ablating YBCO and BaZrO_3 . This yield BaZrO_3 nano-particles and extra c-axis dislocations
 - use of small but non-zero RE ion size variance, mixed RE compositions which produce random defects
 - use of mixed RE's which contain Y and Sm which produce random point defects and correlated defects
 - lower growth temperature of buffer producing surface particles which cause a-b planes to tilt and result in low angle grain boundaries parallel to 'c'